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H. F. Arnoldus and Thomas F. George

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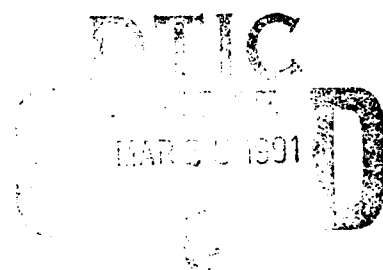
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HEISENBERG APPROACH TO PHOTON EMISSION NEAR A PHASE CONJUGATOR

Henk F. Arnoldus

Department of Physics

Mendel Hall

Villanova University

Villanova, Pennsylvania 19085

and

Thomas F. George

Departments of Chemistry and Physics & Astronomy

239 Fronczak Hall

State University of New York at Buffalo

Buffalo, New York 14260

Abstract

An expression for the emitted fluorescence radiation by an atom near a phase conjugator is derived from a plane-wave expansion of the electric field operator and Heisenberg's equation of motion for the annihilation operator. The result is compared to a solution which was found previously, based on the classical Maxwell equations. It is shown that both theories yield the same expression for the field in the radiation zone, in the limit of a transparent medium. This confirms the correctness of either approach to the problem of optical phase conjugation of atomic radiation.

I. INTRODUCTION

In a previous paper[1] we have studied the behavior of an atom near the surface of a four-wave mixing phase conjugator (PC). We calculated the fluorescent photon emission rate by solving Maxwell's equations for a dipole μ near a PC. The electric field $\underline{E}(\underline{r},t)$ was interpreted as a quantum operator field, with the argument that the classical Maxwell equations must be identical in form to the (quantum) Heisenberg equations of motion for the operator fields. Nevertheless, questions can be raised as to whether the identification of Maxwell's equations with Heisenberg's equations can be justified for the problem under consideration. In particular, the pump beams (with frequency $\bar{\omega}$) for the four-wave mixing process are taken into account parametrically, and are represented by classical plane waves. This leads to factors of $\exp(-2i\bar{\omega}t)$ in the expression for the fluorescence radiation field, rather than $a(t)^2$, with $a(t)$ the annihilation operator for a photon in the pump beam. In addition, our results are in conflict with the results of Hendriks and Nienhuis,[2,3] who did not find the terms proportional to $\underline{\mu}(t)^{(-)}$ (raising part) in the expression for the fluorescence field.

In this paper we solve the Heisenberg equation for the annihilation operator $a_{\underline{k}\sigma}(t)$ for a photon with wave vector $\underline{k}$ and polarization σ (either s = surface polarized or p = plane polarized). The solution is applied to evaluate the fluorescent radiation field int he far zone. These calculations are completely independent from our previous method, and the results can be used to verify the consistency of our approach.

II. ELECTRIC FIELD

The electric-field operator $\underline{E}(\underline{r},t)$ can be represented as a sum of polarized plane waves. In the region $z > 0$ (above the PC, where the atom is),

we have incident (inc) waves with wave vector $\underline{k}$ and polarization σ. These waves gives rise to specularly-reflected (r) waves, and to phase-conjugated (pc) waves which travel in the direction of $-\underline{k}$. In addition, there are transmitted (t) waves which have their origin in waves which are incident on the medium from the other side of the PC. The four-wave mixer produces also a nonlinear (nℓ) wave in $z > 0$.

The amplitude of each of the generated waves is related to the amplitude of the corresponding incident wave by a Fresnel coefficient. The electric field is explicitly,

$$
\underline{E}(\underline{r},t) = \sum_{\underline{k}\sigma}' \sqrt{\frac{\hbar\omega}{2\epsilon_o V}} \left[a_{\underline{k}\sigma}(t) \left\{ \underline{e}_{\underline{k}\sigma}\, e^{i\underline{k}\cdot\underline{r}} + R_{\underline{k}\sigma}\underline{e}_{\underline{k}_r\sigma} e^{i\underline{k}_r\cdot\underline{r}} \right\} \right.
$$

$$
+\, a^{\dagger}_{\underline{k}\sigma}(t)e^{-2i\bar\omega t}\, P^{*}_{\underline{k}\sigma}\underline{e}_{-\underline{k}_{pc}\sigma} e^{-i\underline{k}_{pc}\cdot\underline{r}} \Big]
$$

$$
+ \sum_{\underline{k}\sigma}'' \sqrt{\frac{\hbar\omega}{2\epsilon_o V}} \left[a_{\underline{k}\sigma}(t) T'_{\underline{k}\sigma}\underline{e}_{\underline{k}\sigma} e^{i\underline{k}\cdot\underline{r}} \right.
$$

$$
+\, a^{\dagger}_{\underline{k}\sigma}(t)e^{-2i\bar\omega t}\, N'^{*}_{\underline{k}\sigma}\underline{e}_{-\underline{k}_{n\ell}\sigma} e^{-i\underline{k}_{n\ell}\cdot\underline{r}} \Big] + \text{H.c.} \quad , \qquad (2.1)
$$

where a prime (double prime) on the summation sign indicates a sum over waves which propagate in the $z < 0$ ($z > 0$) direction only. The unit polarization vectors $\underline{e}_{\underline{k}_i\sigma}$ and the wave vectors $\underline{k}_i$ are defined in Appendix A. The Heisenberg operator $a_{\underline{k}\sigma}(t)$ is the annihilation operator for a photon which

is incident on the PC, either from $z > 0$ or from the other side (back port) of the medium. We shall take the Schrödinger picture and Heisenberg picture to coincide at $t = 0$.

III. EQUATION OF MOTION

The only unknown in expression (2.1) for $\underline{E}(\underline{r},t)$ is the annihilation operator $a_{\underline{k}\sigma}(t)$ for $t > 0$. Its equation of motion is

$$i\hbar \frac{d}{dt} a_{\underline{k}\sigma}(t) = [a_{\underline{k}\sigma}(t), H] \quad , \tag{3.1}$$

with initial condition $a_{\underline{k}\sigma}(0) = a_{\underline{k}\sigma}$. The Hamiltonian H can be written as

$$H = H_a + H_r + H_{ar} \quad , \tag{3.2}$$

with

$$H_r = \sum_{\underline{k}\sigma} \hbar\omega a_{\underline{k}\sigma}^{+} a_{\underline{k}\sigma} \tag{3.3}$$

for the Hamiltonian of the radiation field, and where $\omega = ck$. The atomic Hamiltonian H_a can remain unspecified because it commutes with $a_{\underline{k}\sigma}(t)$. For the interaction, we take

$$H_{ar} = -\underline{\mu}\cdot\underline{E}(\underline{h},0) \quad , \tag{3.4}$$

in terms of the atomic dipole moment operator μ and the position $\underline{h} = h\underline{e}_z$ of the atom. With Eqs. (3.3) and (3.4) the equation of motion becomes

$$i\hbar \frac{d}{dt} a_{\underline{k}\sigma}(t) = \hbar\omega \, a_{\underline{k}\sigma}(t) - \underline{\mu}(t) \cdot [a_{\underline{k}\sigma}(t), \underline{E}(\underline{h},t)] \quad , \qquad (3.5)$$

where $\underline{E}(\underline{h},t)$ follows from Eq. (2.1) with $\underline{r} = \underline{h}$.

IV. SOLUTION

An integral of Eq. (3.5) is

$$a_{\underline{k}\sigma}(t) = e^{-i\omega t}\{a_{\underline{k}\sigma} + \frac{i}{\hbar}\int_0^t dt' \, e^{i\omega t'} \underline{\mu}(t') \cdot [a_{\underline{k}\sigma}(t'), \underline{E}(\underline{h},t')]\} \quad . \quad (4.1)$$

When we subsitute Eq. (2.1) for $\underline{E}(\underline{h},t')$, then it appears that many terms in the integrand oscillate at optical frequencies, and these terms average out to zero on a time scale of an optical cycle. The time dependence of the annihilation operator would be $a_{\underline{k}\sigma}(t) = a_{\underline{k}\sigma}\exp(-i\omega t)$ in free evolution, which contains only a single positive frequency. When the interaction is taken into account, $a_{\underline{k}\sigma}(t)$ acquires a spectral width around the frequency ω, but it is still a positive-frequency operator. Similarly, when we split the dipole moment into a positive and a negative frequency part as

$$\underline{\mu}(t) = \underline{\mu}(t)^{(+)} + \underline{\mu}(t)^{(-)} \quad , \qquad (4.2)$$

then $\underline{\mu}(t)^{(+)} = \underline{\mu}^{(+)}\exp(-i\omega_o t)$ in free evolution (for a two-level atom with transition frequency ω_o). The third time dependence enters as $\exp(\pm 2i\bar{\omega}t)$. Then we drop all terms in the integrand which oscillate with optical

frequencies, and retain the terms which oscillate with the difference of two optical frequencies. Furthermore, we notice that the first term on the right-hand side of Eq. (4.1) yields the vacuum field $\underset{\sim}{E}_v(\underset{\sim}{r},t)$.

Therefore, we can write

$$\underset{\sim}{E}(\underset{\sim}{r},t) = \underset{\sim}{E}_v(\underset{\sim}{r},t) + \underset{\sim}{E}_s(\underset{\sim}{r},t) \quad , \tag{4.3}$$

where $\underset{\sim}{E}_v(\underset{\sim}{r},t)$ is given by Eq. (2.1) with $a_{k\sigma}(t) \rightarrow a_{k\sigma}\exp(-i\omega t)$. The source field $\underset{\sim}{E}_s(\underset{\sim}{r},t)$ follows from the second term on the right-hand side of Eq. (4.1). We find explicitly,

$$\underset{\sim}{E}_s(\underset{\sim}{r},t) = \frac{i}{2\epsilon_o V} \sum_{k\sigma}' \omega\, e^{-i\omega t}\, b_{k\sigma}(t) \left\{ \underset{\sim}{e}_{k\sigma} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} + R_{k\sigma} \underset{\sim}{e}_{-k_r\sigma} e^{i\underset{\sim}{k}_r\cdot\underset{\sim}{r}} \right\}$$

$$- \frac{i}{2\epsilon_o V} \left(\sum_{k\sigma}' \omega\, e^{-i(\omega-2\bar\omega)t}\, b_{k\sigma}(t) P_{k\sigma} \underset{\sim}{e}_{-k_{pc}\sigma} e^{i\underset{\sim}{k}_{pc}\cdot\underset{\sim}{r}} \right)^\dagger$$

$$+ \frac{i}{2\epsilon_o V} \sum_{k\sigma}'' \omega\, e^{-i\omega t}\, b'_{k\sigma}(t) T'_{k\sigma} \underset{\sim}{e}_{k\sigma} e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}}$$

$$- \frac{i}{2\epsilon_o V} \left(\sum_{k\sigma}'' \omega\, e^{-i(\omega-2\bar\omega)t}\, b'_{k\sigma}(t) N'_{k\sigma} \underset{\sim}{e}_{-k_{n\ell}\sigma} e^{i\underset{\sim}{k}_{n\ell}\cdot\underset{\sim}{r}} \right)^\dagger + \text{H.c.} \quad , \tag{4.4}$$

in terms of the operators

$$b_{k\sigma}(t) = \int_0^t dt'\, e^{i\omega t'}\, \underset{\sim}{\mu}(t')^{(+)}\cdot\{\underset{\sim}{e}_{k\sigma} e^{-i\underset{\sim}{k}\cdot\underset{\sim}{h}} + R^*_{k\sigma}\underset{\sim}{e}_{-k_r\sigma} e^{-i\underset{\sim}{k}_r\cdot\underset{\sim}{h}}\}$$

$$+ \int_0^t dt' \, e^{i(\omega - 2\bar{\omega})t'} \, \underline{\mu}(t')^{(-)} \cdot (P^*_{\underline{k}\sigma} \, \underline{e}_{\underline{k}_{pc}\sigma} e^{-i\underline{k}_{pc}\cdot\underline{h}}) \quad , \quad k_z < 0 \quad ,$$

$$(4.5)$$

$$b'_{\underline{k}\sigma}(t) = \int_0^t dt' \, e^{i\omega t'} \, \underline{\mu}(t')^{(+)} \cdot (T'^*_{\underline{k}\sigma} \underline{e}_{-\underline{k}\sigma} e^{-i\underline{k}\cdot\underline{h}})$$

$$+ \int_0^t dt' \, e^{i(\omega - 2\bar{\omega})t'} \, \underline{\mu}(t')^{(-)} \cdot (N'^*_{\underline{k}\sigma} \, \underline{e}_{\underline{k}_{n\ell}\sigma} e^{-i\underline{k}_{n\ell}\cdot\underline{h}}) \quad , \quad k_z > 0 \quad .$$

$$(4.6)$$

V. ASYMPTOTIC EXPANSION

In order to simplify the solution (4.4) for $\underline{E}_s(\underline{r},t)$ we consider its value in the radiation zone ($r \to \infty$ and $z > 0$). The operators $b_{\underline{k}\sigma}(t)$ and $b'_{\underline{k}\sigma}(t)$ are independent of $\underline{r}$, so that all $\underline{r}$-dependent factors appear in the form $\exp(i\underline{k}_\alpha \cdot \underline{r})$. These exponentials are multiplied by a function of $\underline{k}_\alpha$, and the result is summed over all values of $\underline{k}_\alpha$. Therefore, all terms in Eq. (4.4) have the generic form $V^{-1} \sum g(\underline{k}_\alpha) \exp(i\underline{k}_\alpha \cdot \underline{r})$. The summation runs either over wave vectors with only positive z-components or over wave vectors with only negative z-components. Changing the summation into an integration gives

$$\frac{1}{V} \sum_{\underline{k}_\alpha} g(\underline{k}_\alpha) e^{i\underline{k}_\alpha \cdot \underline{r}} = \frac{1}{8\pi^3} \int_0^\infty dk_\alpha k_\alpha^2 \int_{\Omega^\pm} d\Omega \, g(\underline{k}_\alpha) e^{i\underline{k}_\alpha \cdot \underline{r}} \quad , \tag{5.1}$$

where the superscript ($\pm$) on the region of solid angle indicates the sign of the z-components of the wave vectors $\underline{k}_\alpha$. Then we can make an asymptotic expansion of the angular integral with the method of stationary phase.[4] The result is

$$\int_{\Omega^{\pm}} d\Omega\; g(\underline{k}_\alpha) e^{i\underline{k}_\alpha \cdot \underline{r}} = \mp\, 2\pi i\, \frac{e^{\pm i k_\alpha r}}{k_\alpha r}\, g(\pm k_\alpha \underline{e}_r) \quad . \tag{5.2}$$

where $\underline{e}_r$ is the radial spherical unit vector which points in the observation direction. This direction will be specified by the spherical angles ϑ and ϕ. The asymptotic expansion effectively filters out the value of $g(\underline{k}_\alpha)$ for $\underline{k}_\alpha = \pm k_\alpha \underline{e}_r$, corresponding to the plane wave $\exp(i\underline{k}_\alpha \cdot \underline{r})$ which travels into the observation direction.

With Eqs. (5.1) and (5.2), the asymptotic expansion of $\underline{E}_s(\underline{r},t)$ is found to be

$$
\underline{E}_s(\underline{r},t) = \frac{-1}{8\pi^2 \epsilon_o r} \times \sum_\sigma \Bigg[\int_0^\infty dk\, k\, e^{-ikr} \big(\omega\, e^{-i\omega t} b_{\underline{k}\sigma}(t)\underline{e}_{\underline{k}\sigma}\big)_{\underline{k}=-k\underline{e}_r}
$$

$$
- \int_0^\infty dk_r\, k_r\, e^{ik_r r} \big(\omega\, e^{-i\omega t} b_{\underline{k}\sigma}(t) R_{\underline{k}\sigma -\underline{k}_r \sigma}\underline{e}_{\underline{k}_r\sigma}\big)_{\underline{k}_r = k_r\underline{e}_r}
$$

$$
+ \left\{ \int_0^\infty dk_{pc}\, k_{pc}\, e^{-ik_{pc} r} \big(\omega\, e^{-i(\omega - 2\bar\omega)t} b_{\underline{k}\sigma}(t) P_{\underline{k}\sigma -\underline{k}_{pc}\sigma}\underline{e}_{\underline{k}_{pc}\sigma}\big)_{\underline{k}_{pc} = -k_{pc}\underline{e}_r} \right\}^\dagger
$$

$$
- \int_0^\infty dk_t\, k_t\, e^{ik_t r} \big(\omega\, e^{-i\omega t} b'_{\underline{k}\sigma}(t) T'_{\underline{k}\sigma -\underline{k}\sigma}\underline{e}_{\underline{k}\sigma}\big)_{\underline{k}_t = k_t\underline{e}_r}
$$

$$
+ \left\{ \int_0^\infty dk_{n\ell}\, k_{n\ell}\, e^{-ik_{n\ell} r} \big(\omega\, e^{-i(\omega - 2\bar\omega)t} b'_{\underline{k}\sigma}(t) \right.
$$

$$
\left. N'_{\underline{k}\sigma -\underline{k}_{n\ell}\sigma}\underline{e}_{\underline{k}_{n\ell}\sigma}\big)_{\underline{k}_{n\ell} = -k_{n\ell}\underline{e}_r} \right\}^\dagger \Bigg] + \text{H.c.} \tag{5.3}
$$

VI. POLARIZATION VECTORS

The polarization vectors $e_{\underset{\sim}{k}_\alpha \sigma}$, evaluated for $\underset{\sim}{k}_\alpha = \pm k_\alpha \underset{\sim}{e}_r$, can be expressed in the spherical unit vectors $\underset{\sim}{e}_\vartheta$ and $\underset{\sim}{e}_\phi$. These calculations are similar (but not identical) to the corresponding calculations in Ref 1 (Sec. IX). Here it is convenient to introduce the notation

$$\underset{\sim}{e}_s^{\pm} = \mp \underset{\sim}{e}_\phi \quad , \qquad \underset{\sim}{e}_p^{\pm} = \underset{\sim}{e}_\vartheta \quad , \tag{6.1}$$

which will enable us to express the results for s-waves and p-waves in a single formula. We find

$$\underset{\sim}{E}_s(\underset{\sim}{r},t) = \frac{1}{8\pi^2 \epsilon_o r} \sum_\sigma [\underset{\sim}{e}_\sigma^- X_{adv,\sigma} + \underset{\sim}{e}_\sigma^+ X_{r,\sigma} + \underset{\sim}{e}_\sigma^- X_{pc,\sigma} + \underset{\sim}{e}_\sigma^+ X_{t,\sigma}$$

$$+ \underset{\sim}{e}_\sigma^- X_{n\ell,\sigma}] + H.c. \quad , \tag{6.2}$$

with

$$X_{adv,\sigma} = -\int_0^\infty dk\, k\omega\, e^{-ikr-i\omega t}\, (b_{\underset{\sim}{k}\sigma}(t))_{\underset{\sim}{k}=-k\underset{\sim}{e}_r} \quad , \tag{6.3}$$

$$X_{r,\sigma} = \int_0^\infty dk_r\, k_r \omega\, e^{ik_r r - i\omega t}\, (b_{\underset{\sim}{k}\sigma}(t) R_{\underset{\sim}{k}\sigma})_{\underset{\sim}{k}_r = k_r \underset{\sim}{e}_r} \quad , \tag{6.4}$$

and similar expressions for $X_{pc,\sigma}$, $X_{t,\sigma}$ and $X_{n\ell,\sigma}$. Apparently, the term proportional to $X_{adv,\sigma}$ is an advanced (non-causal) contribution to $\underset{\sim}{E}_s$, corresponding to incoming spherical waves. The other four terms are retarded (causal) solutions, and they have the form of outgoing spherical waves.

Next we substitute expressions (4.5) and (4.6) for $b_{k\sigma}(t)$ and $b'_{k\sigma}(t)$, respectively, into the results for the $X_{\alpha,\sigma}$'s. Then we evaluate the polarization vectors in $b_{k\sigma}(t)$ and $b'_{k\sigma}(t)$ at the indicated wave vectors. After lengthy calculations we find

$$X_{r,\sigma} = \int_0^\infty dk\ k\omega R_\sigma \times [e^{-i\omega(t-\tau-r/c)} \int_0^t dt'\ e^{i\omega t'}\ \underline{\epsilon}_\sigma \cdot \underline{\mu}(t')^{(+)}$$

$$+ R_\sigma^* e^{-i\omega(t+\tau-r/c)} \int_0^t dt'\ e^{i\omega t'}\ \underline{e}_\sigma^+ \cdot \underline{\mu}(t')^{(+)}$$

$$+ P_\sigma^* e^{-i\omega(t-\tau-r/c)} \int_0^t dt'\ e^{i(\omega-2\bar\omega)t'} \underline{\epsilon}_\sigma \cdot \underline{\mu}(t')^{(-)}] \quad , \qquad (6.5)$$

and four other similar expressions. Here we introduced the abbreviations

$$\underline{\epsilon}_s^+ = -\underline{e}_\phi \quad , \qquad \underline{\epsilon}_p^+ = -\underline{e}_\theta - 2\sin\theta\underline{e}_z \quad , \qquad (6.6)$$

$$\tau = \frac{h}{c}\cos\theta \quad , \qquad (6.7)$$

and all Fresnel coefficients, like P_σ, R_σ, etc., are evaluated at frequency ω and an angle of incidence equal to the polar angle (θ) of observation.

VII. TWO-LEVEL ATOM

So far we have not made any assumption about the atom and its dipole moment $\underline{\mu}(t)$. In order to evaluate the expression for $\underline{E}_s(\underline{r},t)$ further, we assume that only two atomic levels, which might be degenerate, are of

relevance. The level separation is $\hbar\omega_0$. In free evolution, the positive and negative frequency part of $\underline{\mu}(t)$ obey the identity

$$\underline{\mu}(t_2)^{(\pm)} = e^{\mp i\omega_0(t_2-t_1)} \underline{\mu}(t_1)^{(\pm)} \quad .$$ (7.1)

We shall use this as an approximation in equations of the type (6.5). Then, for instance, Eq. (6.5) can be written as

$$\overset{\cdot}{X}_{r,\sigma} = \int_0^\infty dk\ k\omega R_\sigma$$

$$\times\ [\underline{\epsilon}_{-\sigma}^+ \cdot \underline{\mu}(t-\tau-r/c)^{(+)}\ e^{-i(\omega-\omega_0)(t-\tau-r/c)} \int_0^t dt'\ e^{i(\omega-\omega_0)t'}$$

$$+\ R_{\sigma-\sigma}^* \underline{\epsilon}^+ \cdot \underline{\mu}(t+\tau-r/c)^{(+)} e^{-i(\omega-\omega_0)(t+\tau-r/c)} \int_0^t dt'\ e^{i(\omega-\omega_0)t'}$$

$$+ P_{\sigma-\sigma}^* \underline{\epsilon}^+ \cdot \underline{\mu}(t-\tau-r/c)^{(-)} e^{-2i\bar{\omega}(t-\tau-r/c)}$$

$$\times\ e^{-i(\omega+\omega_0-2\bar{\omega})(t-\tau-r/c)} \int_0^t dt'\ e^{i(\omega+\omega_0-2\bar{\omega})t'}\ ] \quad .$$ (7.2)

In every term, both the exponential in the integrand and the exponential in front of the integral have the same frequency. When the distance r is much larger than a wavelength and the retardation r/c is much larger than an optical cycle, then the integrals can be approximated by[5]

$$e^{-i\omega_\alpha(t\pm\tau-r/c)} \int_0^t dt'\ e^{i\omega_\alpha t'} = 2\pi\ \delta(\omega_\alpha) \quad .$$ (7.3)

In writing out all $X_{\alpha,\sigma}$ as in Eq. (7.2), it can be shown that every integral can be written as in Eq. (7.3). The only exception is $X_{adv,\sigma}$, which contains the integral of Eq. (7.3) with r replaced by -r. This makes the right-hand side zero, instead of $2\pi\delta(\omega_\alpha)$, as shown in Ref. 5. Therefore,

$$X_{adv,\sigma} = 0 \quad , \tag{7.4}$$

as it should. Then we set $k = \omega/c$ and carry out the integrations over ω. This gives, for instance,

$$X_{r,\sigma} = \frac{2\pi\omega_o^2}{c^2} \left[R_\sigma \epsilon_{-\sigma}^+ \cdot \mu(t-\tau-r/c)^{(+)} + |R_\sigma|^2 e_{-\sigma}^+ \cdot \mu(t+\tau-r/c)^{(+)} \right.$$

$$\left. + \bar{R}_\sigma \bar{P}_\sigma^* \epsilon_{-\sigma}^+ \cdot \mu(t-\tau-r/c)^{(-)} e^{-2i\bar{\omega}(t-\tau-r/c)} \right] \quad , \tag{7.5}$$

where

$$P_\sigma = P_\sigma(\omega_o, y) \quad , \tag{7.6}$$

$$\bar{P}_\sigma = P_\sigma(2\bar{\omega}-\omega_o, \theta) \quad , \tag{7.7}$$

and similarly for other Fresnel coefficients.

VIII. TOTAL SOURCE FIELD

The total field $E_s(r,t)$ has four contributions of the form (7.5), according to Eq. (6.2). We set $t - r/c \to t$, and introduce the polarization vectors (without the $\pm$ superscripts)

$$\underset{\sim}{e}_s = \underset{\sim}{e}_\phi \quad , \qquad \underset{\sim}{e}_p = \underset{\sim}{e}_\theta \quad , \tag{8.1}$$

$$\underset{\sim}{\epsilon}_s = \underset{\sim}{e}_\phi \quad , \qquad \underset{\sim}{\epsilon}_p = -\underset{\sim}{e}_\theta - 2\sin\theta \underset{\sim}{e}_z \quad . \tag{8.2}$$

Then we define the parameters

$$\gamma_\sigma^a = |R_\sigma|^2 + |T_\sigma'|^2 - |\bar{P}_\sigma|^2 - |\bar{N}_\sigma'|^2 \quad , \tag{8.3}$$

$$\gamma_\sigma^i = \bar{R}_\sigma \bar{P}_\sigma^* - R_\sigma P_\sigma^* + \bar{T}_\sigma' \bar{N}_\sigma'^* - T_\sigma' N_\sigma'^* \quad , \tag{8.4}$$

which will depend on the angle θ, in general. When we group together all terms with $\underset{\sim}{\mu}(t)^{(+)}$ and all terms with $\underset{\sim}{\mu}(t)^{(-)}$, then the field assumes the remarkably simple form

$$\underset{\sim}{E}_s(\underset{\sim}{r},t) = \frac{\omega_o^2 e^{-i\omega_o\tau}}{4\pi\epsilon_o c^2 r} \sum_\sigma \underset{\sim}{e}_\sigma \{\underset{\sim}{a}_\sigma^+ \cdot \underset{\sim}{\mu}(t)^{(+)} + e^{-2i\bar{\omega}t}\underset{\sim}{a}_\sigma^- \cdot \underset{\sim}{\mu}(t)^{(-)}\} + \text{H.c.} \quad , \tag{8.5}$$

in terms of the polarization-like vectors

$$\underset{\sim}{a}_\sigma^+ = \gamma_\sigma^a \underset{\sim}{e}_\sigma + e^{2i\omega_o\tau} R_\sigma \underset{\sim}{\epsilon}_\sigma \quad , \tag{8.6}$$

$$\underset{\sim}{a}_\sigma^- = -P_\sigma^* \underset{\sim}{e}_\sigma + e^{2i\omega_o\tau} \gamma_\sigma^i \underset{\sim}{\epsilon}_\sigma \quad . \tag{8.7}$$

Alternatively, we can group the terms as

$$\underline{E}_s(\underline{r},t) = \frac{\omega_o^2 e^{-i\omega_o \tau}}{4\pi\epsilon_o c^2 r} \{(\underline{M}_\theta \cdot \underline{e}_\theta)\underline{e}_\theta + (\underline{M}_\phi \cdot \underline{e}_\phi)\underline{e}_\phi\} + H.c. \quad , \tag{8.8}$$

with

$$\underline{M}_\theta = \gamma_p^a \underline{\mu}(t)^{(+)} + e^{2i\omega_o \tau} R_p \underline{\mu}'(t)^{(+)}$$

$$- e^{-2i\bar{\omega}t}(P_p^* \underline{\mu}(t)^{(-)} - e^{2i\omega_o \tau} \gamma_p^i \underline{\mu}'(t)^{(-)}) \quad , \tag{8.9}$$

$$\underline{M}_\phi = \gamma_s^a \underline{\mu}(t)^{(+)} - e^{2i\omega_o \tau} R_s \underline{\mu}'(t)^{(+)}$$

$$- e^{-2i\bar{\omega}t}(P_s^* \underline{\mu}(t)^{(-)} + e^{2i\omega_o \tau} \gamma_s^i \underline{\mu}'(t)^{(-)}) \quad . \tag{8.10}$$

The mirror dipole $\underline{\mu}'$ is defined as

$$\underline{\mu}'(t) = \underline{\mu}_\perp(t) - \underline{\mu}_\parallel(t) \quad , \tag{8.11}$$

in terms of the perpendicular and parallel components of $\underline{\mu}(t)$ with respect to the surface $z = 0$. Expressions (8.8) - (8.10) would be identical to our earlier results (12.5) - (12.7) from Ref.1, if the parameters γ_σ^a and γ_σ^i would be $\gamma_\sigma^a = 1$ and $\gamma_\sigma^i = 0$.

IX. SPECIAL CASES

We have not used any of the properties of the Fresnel coefficients in the derivation of the results of the previous section. These coefficients can

be calculated explicitly,[7] but the result is very complicated. Therefore, we
consider two limiting cases of practical interest.

A. Dielectric Layer

When we turn off the pump beams, then the medium becomes an ordinary
dielectric. Therefore,

$$P_\sigma - N_\sigma - 0 \quad , \tag{9.1}$$

which gives

$$\gamma_\sigma^i - 0 \quad . \tag{9.2}$$

The non-zero Fresnel coefficients are related by

$$|T_\sigma|^2 + |R_\sigma^2| - 1 \quad , \tag{9.3}$$

so that

$$\gamma_\sigma^a - 1 \quad . \tag{9.4}$$

This shows that for a dielectric layer, the results (8.8) - (8.10) are
identical to those in Ref.1. Notice that the term which is proportional to
$\underline{\mu}(t)^{(-)}$ in Eq. (8.5) disappears in this limit.

B. <u>Transparent PC</u>

When the dielectric constant equals unity, the specular waves vanish. This gives

$$R_\sigma = N_\sigma = 0 \quad . \tag{9.5}$$

The non-zero Fresnel coefficients are now related by[7]

$$|T_\sigma|^2 - |P_\sigma|^2 = 1 \quad , \tag{9.6}$$

which holds for any polarization, angle of incidence and frequency. Therefore, we find again

$$\gamma_\sigma^a = 1 \quad , \qquad \gamma_\sigma^i = 0 \quad . \tag{9.7}$$

X. <u>CONCLUSION</u>

We have derived an expression for the fluorescence radiation field which is emitted by an atomic dipole near the surface of a PC. The starting point was the standard plane-wave expansion of the electric field in terms of annihilation and creation operators. Then we solved the Heisenberg equation of motion for the annihilation operator. This gives rise to two contributions to the electric field: the vacuum field $\underline{E}_v$ and the source field $\underline{E}_s$, which is generated by the dipole. The form of $\underline{E}_v$ follows trivially from the choice of H_r, but the form of $\underline{E}_s$ depends on the choice of interaction Hamiltonian and the structure of $\underline{E}_v$ (which equals $\underline{E}(\underline{r},0)$). We have shown that $\underline{E}_s$ is identical to the solution of Maxwell's equations, as found previously, provided that

$$\gamma_\sigma^a = 1 \quad , \qquad \gamma_\sigma^i = 0 \quad . \tag{10.1}$$

These parameters do not appear in the solution of Maxwell's equations, which indicates that both approaches are independent indeed. The parameters γ_σ^a and γ_σ^i are determined by the Fresnel reflection and transmission coefficients for a plane wave, and they depend on the polarization, frequency and angle of incidence. The general form of the Fresnel coefficients is extremely complicated, which prohibits the verification of Eq. (10.1) for the most general case. We have shown, however, that for a dielectric layer and for a transparent medium the relations in Eq. (10.1) are satisfied, which covers most cases of practical interest.

ACKNOWLEDGMENT

APPENDIX

Here we summarize the expressions for the various wave vectors in Eq. (2.1) and the phase conventions for the unit polarization vectors. Any wave can be decomposed as

$$\underset{\sim}{k}_i = \underset{\sim}{k}_{\parallel} + k_{i,z} \, \underset{\sim}{e}_z \quad , \tag{A.1}$$

and any $\underset{\sim}{k}_i$ in Eq. (2.1) which corresponds to a given incident wave must have the same parallel component $\underset{\sim}{k}_{\parallel}$ with respect to the surface of the medium. The polarization vectors for s-waves and p-waves are chosen as

$$\underset{\sim}{e}_{\underset{\sim}{k}_i} s = \frac{1}{k_{\parallel}} \, \underset{\sim}{k}_{\parallel} \times \underset{\sim}{e}_z \quad , \tag{A.2}$$

$$\underset{\sim}{e}_{k_i p} = \frac{1}{k_i} \, \underset{\sim}{k}_i \times \underset{\sim}{e}_{\underset{\sim}{k}_i} s \quad , \tag{A.3}$$

respectively. An incident wave from $z > 0$, and with wave vector $\underset{\sim}{k} = k_{\parallel} + k_z \underset{\sim}{e}_z$ generates a specular (r) wave and a phase-conjugated (pc) wave. The z-components of their wave vectors are determined by the dispersion relation, and found to be

$$k_{r,z} = - k_z \quad , \tag{A.4}$$

$$k_{pc,z} = - \sqrt{\rho^2 k^2 - k_{\parallel}^2} \quad , \tag{A.5}$$

in terms of

$$\rho = (2\bar{\omega} - \omega)/\omega \quad . \tag{A.6}$$

A wave with wave vector $\underline{k}$ which is incident on the layer from $z < 0$, generates a transmitted (t) wave in $z > 0$ with wave vector $\underline{k}$. In addition, a nonlinear (nℓ) wave is produced in $z > 0$, which has the same frequency shift with respect to the incident wave as the pc-wave.

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February 11, 1991

Professor Roger W. Anderson
Department of Chemistry
University of California
Santa Cruz, CA 95064

Dear Roger:

Here are some papers to be listed in the upcoming issue #52 of Molecular Dynamics News:

Phase-Conjugated Fluorescence

Phys. Rev. A
Henk F. Arnoldus and Thomas F. George*
Departments of Chemistry and Physics, SUNY-Buffalo, Buffalo, NY 14260
Fluorescent emission by an atom near a phase conjugator (PC) based on four-wave mixing is studied from first principles, where the Maxwell-Heisenberg equations are solved for the radiation field, and with an asymptotic expansion an expression is derived for the field in the far zone. It is pointed out that an atom in its ground state polarizes the nonlinear medium of the PC, which subsequently can emit spontaneously two photons, and an absorption-emission-absorption process by the atom then produces a fluorescent photon, together with a spontaneous excitation of the atom.

Critical Behavior in Magnetic Superlattices

Journal of Magnetism and Magnetic Materials
T. Hai, Z. Y. Li, D. L. Lin and Thomas F. George*
Departments of Chemistry and Physics, SUNY-Buffalo, Buffalo, NY 14260
Based on the effective field theory with correlations, the critical behavior of a magnetic superlattice consisting of two different ferromagnets is examined, where a simple cubic Ising model with nearest-neighbor coupling is assumed. It is found that there exists a critical value for the interface exchange coupling above which the interface magnetism appears.

Interface-Phonon-Mediated Magnetopolaronic Effect on Impurity Transition Energies in Quantum Wells

Phys. Rev. B

D. L. Lin, R. Chen and Thomas F. George*

Departments of Chemistry and Physics, SUNY-Buffalo, Buffalo, NY 14260

Electron interactions with interface phonon modes and strictly-confined bulk phonon modes are considered for the first time to calculate the resonant magnetopolaronic $1s$-$2p_+$ transition energy of a hydrogenic impurity in the quantum well of a double heterostructure, where an interaction gap is predicted between the bulk LO and TO frequences, in contrast to the Fröhlich-type interaction. The results are in good agreement with recent experimental data.

Sincerely,

Thomas F. George

Professor John Baldeschwieler
Department of Chemistry
California Inst. of Technology
Pasadena, CA 91125

Professor John Eyler
Department of Chemistry
University of Florida
Gainesville, FL 32611

Dr. Sylvia Johnson
SRI International
333 Ravenswood Avenue
Menlo Park, CA 94025

Professor Paul Barbara
Department of Chemistry
University of Minnesota
Minneapolis, MN 55455-0431

Professor James Garvey
Department of Chemistry
State University of New York
Buffalo, NY 14214

Dr. Zakya Kafafi
Code 6551
Naval Research Laboratory
Washington, DC 20375-5000

Dr. Duncan Brown
Advanced Technology Materials
520-D Danury Rd.
New Milford, CT 06776

Professor Steven George
Department of Chemistry
Stanford University
Stanford, CA 94305

Professor Larry Kesmodel
Department of Physics
Indiana University
Bloomington, IN 47403

Professor Stanley Bruckenstein
Department of Chemistry
State University of New York
Buffalo, NY 14214

Professor Tom George
Dept. of Chemistry and Physics
State Universety of New York
Buffalo, NY 14260

Professor Max Lagally
Dept. Metal. & Min. Engineerin
University of Wisconsin
Madison, WI 53706

Professor Carolyn Cassady
Department of Chemistry
Miami University
Oxford, OH 45056

Dr. Robert Hamers
IBM T.J. Watson Research Center
P.O. Box 218
Yorktown Heights, N Y 10598

Dr. Stephen Lieberman
Code 522
Naval Ocean Systems Center
San Diego, CA 92152

Professor R.P.H. Chang
Dept. Matls. Sci. & Engineering
Northwestern University
Evanston, IL 60208

Professor Charles Harris
Department of Chemistry
University of California
Berkeley, CA 94720

Professor M.C. Lin
Department of Chemistry
Emory University
Atlanta, GA 30322

Professor Frank DiSalvo
Department of Chemistry
Cornell University
Ithaca, NY 14853

Professor John Hemminger
Department of Chemistry
University of California
Irvine, CA 92717

Professor Fred McLafferty
Department of Chemistry
Cornell University
Ithaca, NY 14853-1301

Dr. James Duncan
Federal Systems Division
Eastman Kodak Company
Rochester, NY 14650-2156

Professor Leonard Interrante
Department of Chemistry
Rensselaer Polytechnic Institute
Troy, NY 12181

Professor Horia Metiu
Department of Chemistry
University of California
Santa Barbara, CA 93106

Professor Arthur Ellis
Department of Chemistry
University of Wisconsin
Madison, WI 53706

Professor Roald Hoffmann
Department of Chemistry
Cornell University
Ithaca, NY 14853

Professor Larry Miller
Department of Chemistry
University of Minnesota
Minneapolis. MN 55455-0431

Professor Mustafa El-Sayed
Department of Chemistry
University of California
Los Angeles, CA 90024

Professor Eugene Irene
Department of Chemistry
University of North Carolina
Chapel Hill, NC 27514

Professor George Morrison
Department of Chemistry
Cornell University
Ithaca, NY 14853

Professor Daniel Neumark
Department of Chemistry
University of California
Berkeley, CA   94720

Professor Robert Whetten
Department of Chemistry
University of California
Los Angeles, CA   90024

Professor David Ramaker
Department of Chemistry
George Washington University
Washington, DC   20052

Professor R. Stanley Williams
Department of Chemistry
University of California
Los Angeles, CA   90024

Dr. Gary Rubloff
IBM T.J. Watson Research Center
P.O. Box 218
Yorktown Heights, NY   10598

Professor Nicholas Winograd
Department of Chemistry
Pennsylvania State University
University Park, PA   16902

Professor Richard Smalley
Department of Chemistry
Rice University
P.O. Box 1892
Houston, TX   77251

Professor Aaron Wold
Department of Chemistry
Brown University
Providence, RI   02912

Professor Gerald Stringfellow
Dept. of Matls. Sci.
     & Engineering
University of Utah
Salt Lake City, UT   84112

Professor Vicki Wysocki
Department of Chemistry
Virginia Commonwealth University
Richmond, VA   23284-2006

Professor Galen Stucky
Department of Chemistry
University of California
Santa Barbara, CA   93106

Professor John Yates
Department of Chemistry
University of Pittsburg
Pittsburg, PA   15260

Professor H. Tachikawa
Department of Chemistry
Jackson State University
Jackson, MI   39217-0510

Professor William Unertl
Lab. for Surface Sci.
     & Technology
University of Maine
Orono, ME   04469

Dr. Terrell Vanderah
Code 3854
Naval Weapons Center
China Lake, CA   93555

Professor John Weaver
Dept. of Chem. Eng. & Mat. Sci.
University of Minnesota
Minneapolis, MN   55455

Professor Brad Weiner
Department of Chemistry
University of Puerto Rico
Rio Piedras, Puerto Rico   00931